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Cosmogenic noble gases in silicate inclusions of iron meteorites: Effects of bulk composition on elemental production rates.

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Abstract

The influence of bulk chemical composition on elemental production rates of neon has been studied experimentally by investigating mineral samples separated from the iron meteorites Four Corners, Landes, Linwood and Zagora. The neon in these samples is purely cosmogenic and its isotopic composition indicates an enhanced production of ^{21}Ne. This is due to the production of additional secondary neutrons caused by a larger multiplicity in meteorites of high-Z bulk chemistry.

Introduction

Meteoroids as meter-sized bodies are irradiated in space by high-energy cosmic-ray particles that penetrate these bodies and interact with its matter. Bauer (1947) and Huntley (1948) independently proposed that measurable amounts of He should be produced in iron meteorites. Paneth *et al.* (1952) showed that this was indeed true and that cosmic-ray produced ^{3}He/^{4}He ratio is in the range 0.2 to 0.3 and were thus very different from atmospheric He. Soon thereafter, also cosmogenic Ne and Ar isotopes were discovered (Reasbeck & Mayne 1955; Gentner & Zähringer 1957). Since than, cosmic-ray produced ("cosmogenic") stable or radioactive reaction products have been studied in great detail to investigate the irradiation history of extraterrestrial matter or the nature of cosmic rays in the past (see *e.g.* Anders 1962; Lal 1972; Reedy *et al.* 1983; Vogt *et al.* 1990).

The production rate of cosmogenic nuclides is dependent on the chemical composition of the target material, the properties of the incident particle irradiation, and the shielding conditions (pre-atmospheric size of the meteoroid and sample's location within the meteoroid). The effects of shielding are mainly caused by the fact that in nuclear reactions initiated by primary cosmic rays secondary particles are also produced. These secondary particles, mostly neutrons of smaller energy, cause further nuclear reactions. Elemental or isotopic nuclide ratios produced by these low-energy particles are different from those of the primary high-energy irradiation. Thus, the shielding of a sample is characterized by elemental or isotopic ratios of specific cosmogenic nuclides produced in different percentages by primary and secondary particles. For example, for shielding corrections of production rates of stony meteorites, cosmogenic ^{22}Ne/^{21}Ne is commonly used (*e.g.* Eugster 1988), or for iron meteorites, the ^{4}He/^{38}Ar ratio (*e.g.* Voshage & Feldmann 1978) is a measure for shielding.

The production rates of cosmogenic nuclides depend twofold on the bulk chemical composition of the meteorite. First, the production of a particular nuclide is given by the concentration-weighted sum of the individual elemental production rates from each target element in the meteorite and, secondly, the elemental production rates themselves depend on it since spectra and intensities of secondary particles inside the meteoroid are influenced by the mean mass number and the mean atomic number of the bulk meteorite. In particular, the multiplicity for the production of secondary neutrons is greater in metal-rich meteorites compared to stony meteorites.

The latter ("matrix") effect was first suggested by Begemann & Schultz (1988). This idea is based on measurements of cosmogenic noble gases in metal and silicates of mesosiderites (Begemann *et al.* 1976). These authors had observed that the concentration of cosmogenic ^{38}Ar produced from Ca in silicates (mainly by secondary neutrons) is larger than expected from the ^{38}Ar produced in metal by the primary irradiation. Begemann and Schultz (1988) show that the production rate ratio of ^{38}Ar from Ca to that of ^{38}Ar from Fe is correlated with the total bulk metal content of mesosiderites. Also, the ^{22}Ne/^{21}Ne ratio in these silicate samples is relatively low. This is explained by an enhanced production of ^{21}Ne via secondary neutrons and the reaction ^{24}Mg(n,α)^{21}Ne.

The matrix effect was also treated theoretically by Michel *et al.* (1990) and Masarik & Reedy (1994) using model calculations which combine spectra of primary and secondary particles derived from Monte Carlo calculations of intra- and internuclear cascades with thin-target cross sections of the underlying nuclear reactions. These calculations show, for example, that the matrix effect is most pronounced in the production rate of ^{21}Ne from Mg, and compared to stony meteorites the ^{21}Ne production is enhanced by a factor of up to two in silicate inclusions of iron meteorites.

We report here measurements of concentration and isotopic composition of neon in silicates separated from three IA iron meteorites with silicate inclusions (Landes, Linwood and Zagora) and from the IB iron Four Corners. This work was carried out to obtain information on the matrix effect in iron meteorites which are more metal-rich than the previously investigated mesosiderites.

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Experimental Methods and Results

Carbon and silicate phases of Landes and Zagora were separated from the meteorite by anodic dissolution of the FeNi. After several ultrasonic treatments and quenching steps with liquid N_2, a further separation of different grain size fractions took place using density and magnetic separation techniques. The Four Corners and Linwood separates were obtained from R S Clarke (Washington DC) and are leftovers from investigations by G P Merill and E P Henderson.

The concentration of the elements Mg, Al, Si, K, Ca, Ti and Fe of selected separates was determined using x-ray fluorescence (XRF) techniques. The isotopic composition and concentrations of noble gases were measured using apparatus and experimental procedures as described by Schultz *et al.* (1991). Results are given in Table 1 together with estimates of the mineral composition of the samples calculated from its chemical composition.

Figure 1 shows isotopic ratios of neon in a 3-isotope plot. Cosmogenic $^{20}Ne/^{22}Ne$ ratios in ordinary chondrites are rather independent of shielding ($^{20}Ne/^{22}Ne = 0.84 \pm 0.02$) and are found in a narrow area (Fig 1; hatched, marked "Chondrites"). If the measured neon is a mixture of cosmogenic gas and a trapped component (solar or planetary gas, atmospheric contamination) the data points would be shifted to higher $^{20}Ne/^{22}Ne$ values. The low $^{20}Ne/^{22}Ne$ values indicate, however, that no significant abundance of trapped Ne is present in spite of trapped Ar, Kr and Xe that is commonly observed in silicate inclusions of iron meteorites (Hintenberger *et al.* 1969; Niemeyer 1979; Crabb 1983; Jentsch & Schultz 1987). The displacement of many data points to higher $^{21}Ne/^{22}Ne$ values is discussed below.

The three feldspar-rich samples are characterized by smaller $^{20}Ne/^{22}Ne$ and $^{21}Ne/^{22}Ne$ ratios. This is due to the relatively high concentration of Na in these samples. Sodium is an element that has a high production rate for ^{22}Ne compared to that of the other two neon isotopes. Smith & Huneke (1975) observed $^{20}Ne/^{22}Ne$ ratios < 0.7 in plagioclase separated from ordinary chondrites. Our feldspar measurement of Zagora is very similar to this value. The two Landes samples with a smaller percentage of feldspar (and thus Na) are located on the mixing line between the "pure" feldspar neon ratio and "normal" cosmogenic neon. Unfortunately, the concentration of Na was not measured directly in these samples because $NaNO_3$ was used to make the tablets for XRF-measurements.

Table 1.

Chemical and mineralogical information on the investigated separates as well as on their concentration and isotopic composition of neon.

Meteorite and sample #	Mg	Al	Si	K	Ca	Ti	Fe	Mg/(Mg+Al+Si)	Mineral composition Opx	Ol	Cpx	Fsp	^{20}Ne	^{21}Ne	^{22}Ne	$^{22}Ne/^{21}Ne$
	[wt %]							[wt.% ratio]	[wt.%]				[in $10^{-8}cm^3STP/g$]			
Four Corners																
401	19.5	1.22	26.5	0.075	2.50	0.13	3.8	0.414	67	12	12	9	348.1	407.9	394.6	0.967
Landes																
I364	22.4	0.11	26.0	0.022	1.20	0.12	4.0	0.462	81	14	5	-	78.5	89.4	91.3	1.021
I344	21.9	0.35	25.7	0.021	1.59	0.13	4.3	0.456	79	14	7	-	75.2	85.5	88.0	1.029
I363	26.9	0.09	23.0	0.016	.78	0.07	3.8	0.539	42	54	4	-	84.8	97.7	98.5	1.008
I355	4.6	9.90	31.2	0.36	1.95	0.07	2.9	0.100	22	-	-	78	15.2	17.1	19.9	1.161
II345	22.2	0.37	25.6	0.027	1.42	0.13	4.3	0.460	78	15	7	-	79.7	91.2	92.8	1.017
II365	23.1	0.23	25.3	0.022	1.14	0.17	4.1	0.474	74	21	5	-	60.5	68.9	70.3	1.021
II366	26.0	0.11	23.7	0.015	0.69	0.08	3.7	0.522	53	44	3	-	89.9	103.2	103.8	1.006
II353	5.4	9.40	31.2	0.33	1.98	0.07	2.6	0.117	26	-	-	74	20.2	22.7	26.4	1.162
III343	21.4	0.44	25.1	0.021	2.38	0.13	5.0	0.456	70	17	13	-	75.3	85.8	87.6	1.021
III347	25.2	0.21	23.7	0.010	1.49	0.10	3.9	0.513	49	43	8	-	86.5	99.8	100.9	1.011
Linwood																
425	11.5	3.49	30.8	0.21	3.23	0.14	2.7	0.252	43	-	13	44	32.3	36.6	36.1	0.987
Zagora																
I329	23.0	0.22	25.4	0.015	1.12	0.13	4.1	0.473	76	19	5	-	91.6	108.2	104.3	0.964
I417	24.4	0.15	24.6	0.015	0.99	0.12	3.9	0.496	65	31	4	-	118.0	139.1	133.5	0.960
I328	22.2	0.24	25.5	0.023	1.21	0.14	4.8	0.464	63	32	5	-	113.7	133.7	129.2	0.966
II322	23.5	0.07	24.3	0.013	0.81	0.11	5.9	0.490	70	27	3	-	110.8	130.2	125.4	0.963
II320	24.3	0.09	24.1	0.017	0.86	0.11	5.1	0.501	64	33	3	-	110.1	129.9	124.0	0.955
II319	23.7	-	23.3	0.026	1.05	0.10	7.1	0.504	59	36	5	-	112.6	132.6	127.5	0.962
II325	25.3	0.08	22.8	0.020	2.54	0.09	4.3	0.525	33	52	15	-	119.6	141.9	135.1	0.952
III386	0.94	11.8	32.8	0.63	1.82	0.05	2.1	0.021	9	-	-	91	20.8	23.1	35.2	1.523

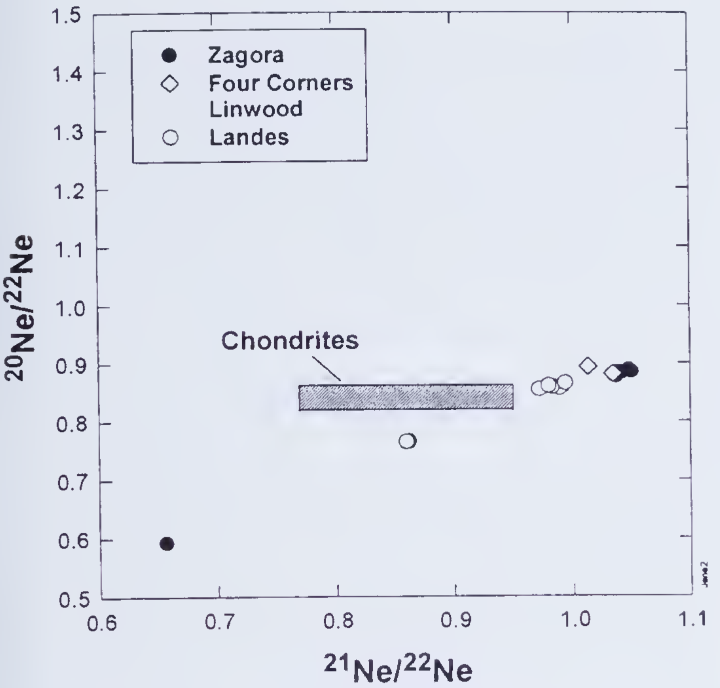

Figure 1. A three-isotope plot of neon showing the position of cosmogenic neon in chondrites (hatched area) and those of analyzed minerals separated from iron meteorites. Trapped components or atmospheric contamination, characterized by high $^{20}Ne/^{22}Ne$ and low $^{21}Ne/^{22}Ne$ values, are not present in appreciable amounts. Data points below the chondritic values are feldspar-rich samples with higher concentrations of sodium that cause higher production rates of ^{22}Ne.

Discussion

An enhanced flux of secondary neutrons in FeNi-rich meteorites will produce lower cosmogenic $^{22}Ne/^{21}Ne$ due to the reaction $^{24}Mg(n,\alpha)^{21}Ne$. To show this effect one must refer to a similar Mg concentration between normal chondritic samples and silicate inclusions of iron meteorites. In addition, the minimum values of cosmogenic $^{22}Ne/^{21}Ne$ for chondrites must be known.

For stony meteorites the shielding effect has been studied on individual samples as well as on samples taken from drill cores of meteorites. Eberhardt *et al.* (1966) found a relation between cosmogenic $^{3}He/^{21}Ne$ and $^{22}Ne/^{21}Ne$ that is widely used for shielding correction of production rates. This relation - originally observed in samples of different chondrites - is shown in Figure 2 together with a number of measurements for samples taken from defined locations in larger meteorites or , as in the case of Kokubunji, taken from different samples of a meteorite shower. A lower limit of observed cosmogenic $^{22}Ne/^{21}Ne$ ratios is about 1.06. This value was also confirmed in measurements of the largest stony meteorite analyzed so far, the Jilin chondrite (Begemann *et al.* 1995). These data are shown in Figure 3. Jilin has had a two-stage exposure history, a short 4π irradiation as a meteoroid and an extended one in 2π geometry (Begemann *et al.* 1985). All data have $^{22}Ne/^{21}Ne > 1.06$ but the measured concentrations of ^{21}Ne vary by a factor of about 6. They do not follow the expected variation of the production rate with shielding (*e.g.* Eugster 1988) as indicated by the heavy curve in Figure 3. The Jilin data clearly show that the lower limit for $^{22}Ne/^{21}Ne$ in chondrites is about 1.06. The calculation of exposure ages using $^{22}Ne/^{21}Ne$ as a shielding indicator may yield a lower limit only if cosmogenic $^{22}Ne/^{21}Ne$ is less than about 1.08.

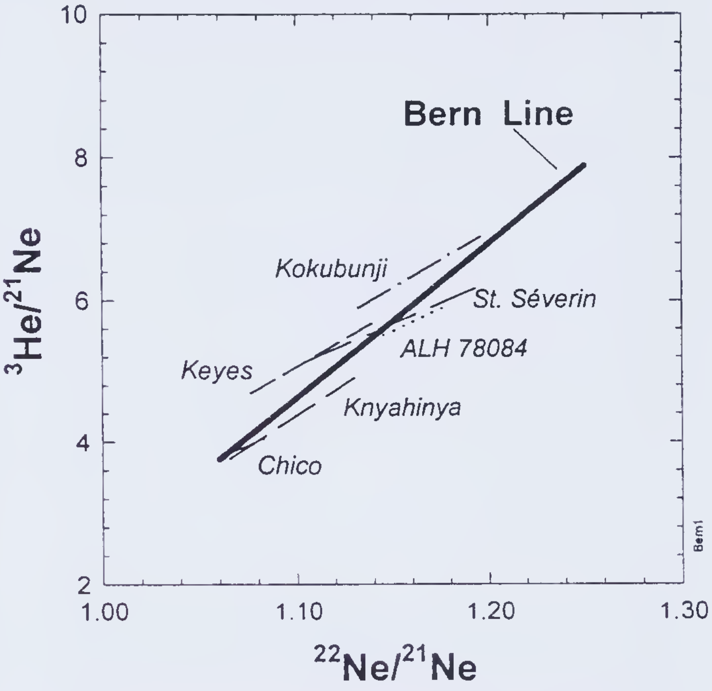

Figure 2. Cosmogenic $^{3}He/^{21}Ne$ versus $^{22}Ne/^{21}Ne$ in chondrites. The Bern-Line (Eberhardt *et al.* 1966) is obtained from measurements of different meteorites. Other lines are measurements of samples taken from defined locations within one meteorite or from individual specimens of a meteorite shower (adapted from Loeken *et al.* 1992; references are given therein).

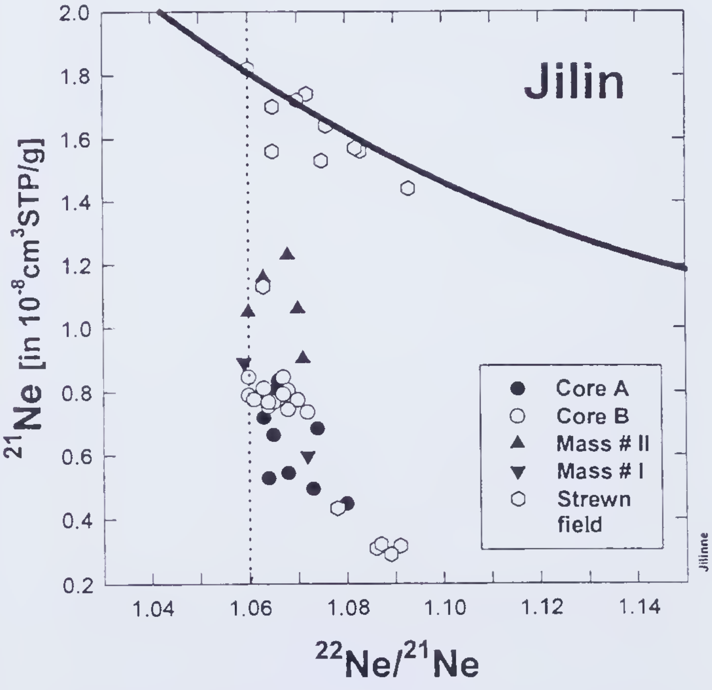

Figure 3. Concentration of cosmogenic ^{21}Ne as function of cosmogenic $^{22}Ne/^{21}Ne$ in samples taken from the Jilin chondrite (Begemann *et al.* 1995). The solid curve is the expected trend of the shielding effect on the production rate of ^{21}Ne (Eugster 1988; normalized to $^{21}Ne = 1.8 \times 10^{-8}$ $cm^{3}STPg$ and $^{22}Ne/^{21}Ne = 1.06$). These measurements show clearly that for $^{22}Ne/^{21}Ne < 1.06$ the predicted relation is not valid and that $^{22}Ne/^{21}Ne = 1.06$ is the lower limit of cosmogenic neon in chondrites.

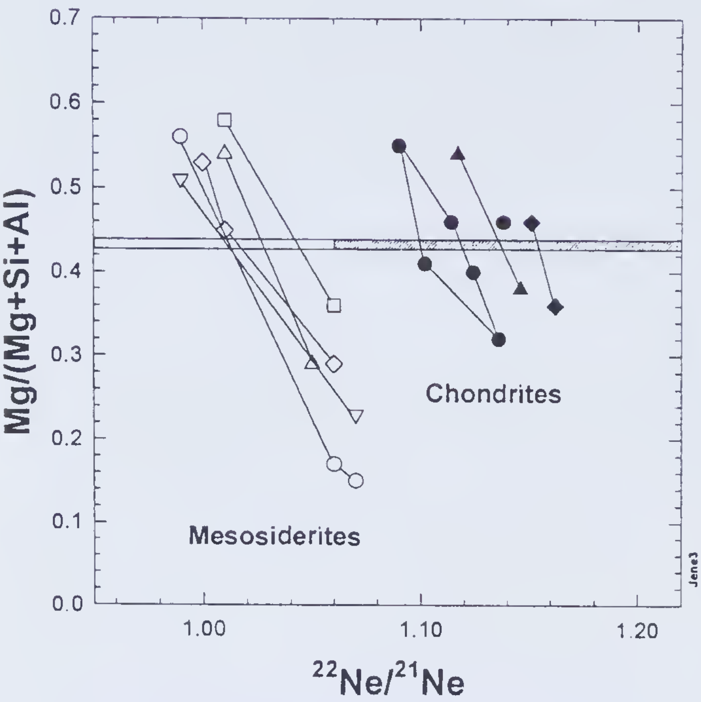

Figure 4. Cosmogenic $^{22}Ne/^{21}Ne$ ratios of silicates from chondrites and mesosiderates that have values smaller than 1.06 (adapted from Begemann & Schultz 1988). Data points are connected when different mineral separates from the same meteorite have been analyzed.

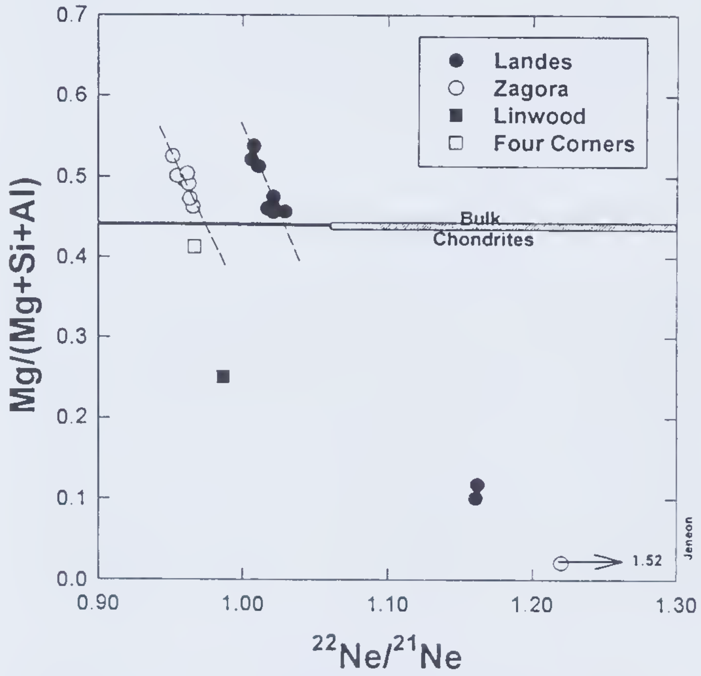

Figure 5. $^{22}Ne/^{21}Ne$ ratios of silicate separates from iron meteorites as function of their Mg/(Mg+Si+Al) ratio. The position of chondrites is shown by the hatched area. For the same chemical composition the silicates of iron meteorites have lower $^{22}Ne/^{21}Ne$ ratios due to an enhanced flux of secondary neutrons. Samples with low Mg concentrations are feldspar-rich with relatively high sodium contents. Their $^{22}Ne/^{21}Ne$ is influenced by higher production rates of ^{22}Ne from Na.

It should also be noted that in the metal phase of iron meteorites, cosmogenic $^{22}Ne/^{21}Ne$ is generally greater than 1.06 (see compilation by Schultz & Kruse 1989).

Figure 4 (adopted from Begemann & Schultz 1988) shows $^{22}Ne/^{21}Ne$ as a function of the Mg(Mg+Si+Al) ratio. More than 95% of cosmogenic neon is produced in chondrites under normal shielding conditions from these three elements, about 80% alone from Mg. There is a small dependence of the $^{22}Ne/^{21}Ne$ on the chemical composition as indicated by the lines connecting measurements of mineral separates of individual meteorites. Referring to the same chemistry [= Mg/(Mg+Si+Al)] the lowest $^{22}Ne/^{21}Ne$ of mesosiderites is about 1.01 and definitely lower than the lower limit for chondrites of 1.06.

Figure 5 shows a similar diagram for the measurements of silicate inclusions from iron meteorites. The mineral separates of Landes and Zagora show a similar dependence of $^{22}Ne/^{21}Ne$ ratios on chemistry as observed for mesosiderites; however, the feldspar samples do not follow this trend because of the ^{22}Ne produced from Na (see above). The $^{22}Ne/^{21}Ne$ - if taken at the chondrite chemical composition - is 1.03 for Landes and 0.97 for Zagora. Those of Linwood and Four Corners cannot be deduced with certainty from this graph because only one sample of each was measured. However, assuming similar relations between chemistry and $^{22}Ne/^{21}Ne$ as observed for Landes and Zagora, the cosmogenic $^{22}Ne/^{21}Ne$ is even smaller than 0.97.

The underlying physical reason for smaller cosmogenic $^{22}Ne/^{21}Ne$ ratios in iron-rich meteorites compared to chondrites is the higher multiplicity for the production of secondary neutrons from high-Z elements like Fe and Ni as compared to Mg, Si or Al (Begemann & Schultz 1988). These additional neutrons enhance the production rate of ^{21}Ne via the reaction $^{24}Mg(n,\alpha)^{21}Ne$ and, thus, lower the cosmogenic $^{22}Ne/^{21}Ne$ ratios.

This effect complicates their use as a shielding indicator of silicates in iron-rich meteorites because $^{22}Ne/^{21}Ne$ depends on the following:

(1) Chemical composition of the analyzed sample, especially the Mg concentration. This effect is seen by the dashed lines in Figure 5.

(2) The chemical composition of the bulk meteorite because a high-Z matrix will change the secondary neutron flux. The difference between individual iron meteorites in Figure 5, after taking into account the chemical composition of the minerals, is presumably partly due to their different percentage of silicates, graphite and troilite.

(3) Shielding as a function of preatmospheric size and location of the analyzed sample.

This complicated relationship makes it difficult to use the cosmogenic $^{22}Ne/^{21}Ne$ in silicates separated from iron rich meteorites as a shielding correction factor of cosmogenic nuclide production rates. Furthermore, model calculations or empirical models describing production rates of cosmogenic nuclides in iron meteorites (*e.g.* Nagai *et al.* 1993) must consider this matrix effect.

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